

## Dependence of nonlocal Gilbert damping on the ferromagnetic layer type in FM/Cu/Pt heterostructures

A. Ghosh, J.F. Sierra, S. Auffret, U. Ebels<sup>1</sup> and W.E. Bailey<sup>2</sup>

<sup>1</sup>*SPINTEC, UMR(8191) CEA / CNRS / UJF / Grenoble INP ; INAC,  
17 rue des Martyrs, 38054 Grenoble Cedex, France*

<sup>2</sup>*Dept. of Applied Physics & Applied Mathematics, Columbia University,  
New York NY 10027, USA*

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We have measured the size effect in nonlocal Gilbert relaxation rate in FM( $t_{FM}$ ) / Cu (5nm) [/ Pt (2nm)] / Al(2nm) heterostructures, FM = { Ni<sub>81</sub>Fe<sub>19</sub>, Co<sub>60</sub>Fe<sub>20</sub>B<sub>20</sub>, pure Co}. Common behavior is observed for three FM layers, where the additional relaxation obeys both a strict inverse power law dependence  $\Delta G = K t^n$ ,  $n = -1.04 \pm 0.06$  and a similar magnitude  $K = 224 \pm 40$  Mhz · nm. As the tested FM layers span an order of magnitude in spin diffusion length  $\lambda_{SDL}$ , the results are in support of spin diffusion, rather than nonlocal resistivity, as the origin of the effect.

The primary materials parameter which describes the temporal response of magnetization  $\mathbf{M}$  to applied fields  $\mathbf{H}$  is the Gilbert damping parameter  $\alpha$ , or relaxation rate  $G = |\gamma|M_s\alpha$ . Understanding of the Gilbert relaxation, particularly in structures of reduced dimension, is an essential question for optimizing the high speed / GHz response of nanoscale magnetic devices.

Experiments over the last decade have established that the Gilbert relaxation of ferromagnetic ultrathin films exhibits a size effect, some component of which is nonlocal. Both  $\alpha(t_{FM}) = \alpha_0 + \alpha'(t_{FM})$  and  $G(t_{FM}) = G_0 + G'(t_{FM})$  increase severalfold with decreasing FM film thickness  $t_{FM}$ , from near-bulk values  $\alpha_0, G_0$  for  $t_{FM} \gtrsim 20$  nm. Moreover, the damping size effect can have a nonlocal contribution responsive to layers or scattering centers removed, through a nonmagnetic (NM) layer, from the precessing FM. Contributed Gilbert relaxation has been seen from other FM layers<sup>1</sup> as well as from heavy-element scattering layers such as Pt.<sup>2</sup>

The nonlocal damping size effect is strongly reminiscent of the electrical resistivity in ferromagnetic ultrathin films. Electrical resistivity  $\rho$  is size-dependent by a similar factor over a similar range of  $t_{FM}$ ; the resistivity  $\rho(t_{FM})$  is similarly nonlocal, dependent upon layers not in direct contact.<sup>3-5</sup> It is *prima facie* plausible that the nonlocal damping and nonlocal electrical resistivity share a common origin in momentum scattering (with relaxation time  $\tau_M$ ) by overlayers. If the nonlocal damping arises from nonlocal scattering  $\tau_M^{-1}$ , however, there should be a marked dependence upon FM layer type. Damping in materials with short spin diffusion length  $\lambda_{SDL}$  is thought to be proportional to  $\tau_M^{-1}$  (ref.<sup>6</sup>); the claim for "resistivity-like" damping has been made explicitly for  $\text{Ni}_{81}\text{Fe}_{19}$  by Ingvarsson<sup>7</sup> et al. For FM with a long  $\lambda_{SDL}$ , on the other hand, relaxation  $G$  is either nearly constant with temperature or "conductivity-like," scaling as  $\tau_M$ .

Interpretation of the nonlocal damping size effect has centered instead on a spin current model<sup>8</sup> advanced by Tserkovnyak et al<sup>9</sup>. An explicit prediction of this model is that the magnitude of the nonlocal Gilbert relaxation rate  $\Delta G$  is only weakly dependent upon the FM layer type. The effect has been calculated<sup>10</sup> as

$$\Delta G = |\gamma|^2 \hbar / 4\pi \left( g_{eff}^{\uparrow\downarrow} / S \right) t_{FM}^{-1} \quad (1)$$

, where the effective spin mixing conductance  $g_{eff}^{\uparrow\downarrow} / S$  is given in units of channels per area. *Ab-initio* calculations predict a very weak materials dependence for the interfacial parameters

$g^{\uparrow\downarrow}/S$ , with  $\pm 10\%$  difference in systems as different as Fe/Au and Co/Cu, and negligible dependence on interfacial mixing.<sup>11</sup>

Individual measurements exist of the spin mixing conductance, through the damping, in FM systems Ni<sub>81</sub>Fe<sub>19</sub><sup>12</sup>, Co<sup>13</sup>, and CoFeB<sup>14</sup>. However, these experiments do not share a common methodology, which makes a numerical comparison of the results problematic, especially given that Gilbert damping estimates are to some extent model-dependent<sup>15</sup>. In our experiments, we have taken care to isolate the nonlocal damping contribution due to Pt overlayers only, controlling for growth effects, interfacial intermixing, and inhomogeneous losses. The only variable in our comparison of nonlocal damping  $\Delta G(t_{FM})$ , to the extent possible, has been the identity of the FM layer.

Gilbert damping  $\alpha$  has been measured through ferromagnetic resonance (FMR) from  $\omega/2\pi = 2\text{-}24$  GHz using a broadband coplanar waveguide (CPW) with broad center conductor width  $w = 400\mu m$ , using field modulation and lock-in detection of the transmitted signal to enhance sensitivity. The Gilbert damping has been separated from inhomogeneous broadening in the films measured using the well-known relation  $\Delta H_{pp}(\omega) = \Delta H_0 + (2/\sqrt{3}) \alpha \omega / |\gamma|$ . We have fit spectra to Lorentzian derivatives with Dysonian components at each frequency, for each film, to extract the linewidth  $\Delta H_{pp}$  and resonance field  $H_{res}$ ;  $\alpha$  has been extracted using linear fits to  $\Delta H(\omega)$ .

For the films, six series of heterostructures were deposited of the form Si/ SiO<sub>2</sub>/ X/ FM( $t_{FM}$ )/ Cu(3nm)/ **Pt(3nm)**/ Al(3nm), FM = { Ni<sub>81</sub>Fe<sub>19</sub> ("Py"), Co<sub>60</sub>Fe<sub>20</sub>B<sub>20</sub> ("CoFeB"), pure Co}, and  $t_{FM} = 2.5, 3.5, 6.0, 10.0, 17.5, 30.0$  nm, for 36 heterostructures included in the study. For each ferromagnetic layer type  $FM$ , one thickness series  $t_{FM}$  was deposited with the Pt overlayer and one thickness series  $t_{FM}$  was deposited without the Pt overlayer. This makes it possible to record the additional damping  $\Delta\alpha(t_{FM})$  introduced by the Pt overlayer alone, independent of size effects present in the FM/Cu layers deposited below. In the case of pure Co, a X=Ta(5nm)/Cu(5nm) underlayer was necessary to stabilize low-linewidth films, otherwise, depositions were carried out directly upon the *in-situ* ion-cleaned substrate.

Field-for-resonance data are presented in Figure 1. The main panel shows  $\omega(H_B^{\parallel})$  data for Ni<sub>81</sub>Fe<sub>19</sub>( $t_{FM}$ ). Note that there is a size effect in  $\omega(H_B^{\parallel})$ : the thinner films have a substantially lower resonance frequency. For  $t_{FM} = 2.5$  nm, the resonance frequency is depressed by  $\sim 5$  GHz from  $\sim 20$  GHz resonance  $H_B \simeq 4$  kOe. The behavior is fitted through

the Kittel relation (lines)  $\omega(H_B^\parallel) = |\gamma| \sqrt{(H_B^\parallel + H_K)(4\pi M_s^{eff} + H_B^\parallel + H_K)}$ , and the inset shows a summary of extracted  $4\pi M_s^{eff}(t_{FM})$  data for the three different FM layers. Samples with (open symbols) and without (closed symbols) Pt overlayers show negligible differences. Linear fits according to  $4\pi M_s^{eff}(t_{FM}) = 4\pi M_s - (2K_s/M_s)t_{FM}^{-1}$  allow the extraction of bulk magnetization  $4\pi M_s$  and surface anisotropy  $K_s$ ; we find  $4\pi M_s^{Py} = 10.7$  kG,  $4\pi M_s^{CoFeB} = 11.8$  kG,  $4\pi M_s^{Co} = 18.3$  kG, and  $K_s^{Py} = 0.69$  erg/cm<sup>2</sup>,  $K_s^{CoFeB} = 0.69$  erg/cm<sup>2</sup>,  $K_s^{Co} = 1.04$  erg/cm<sup>2</sup>. The value of  $g_L/2 = |\gamma|/(e/mc)$ ,  $|\gamma| = 2\pi \cdot (2.799 \text{ Mhz/Oe}) \cdot (g_L/2)$  is found from the Kittel fits subject to this choice, yielding  $g_L^{Py} = 2.09$ ,  $g_L^{CoFeB} = 2.07$ ,  $g_L^{Co} = 2.15$ . The  $4\pi M_s$  and  $g_L$  values, taken to be size-independent, are in good agreement with bulk values.

FMR linewidth as a function of frequency  $\Delta H_{pp}(\omega)$  is plotted in Figure 2. The data for Py show a near-proportionality, with negligible inhomogeneous component  $\Delta H_0 \leq 4$  Oe even for the the thinnest layers, facilitating the extraction of intrinsic damping parameter  $\alpha$ . The size effect in  $\alpha(t_{FM})$  accounts for an increase by a factor of  $\sim 3$ , from  $\alpha_0^{Py} = 0.0067$  ( $G_0^{Py} = 105$  Mhz) for the thickest films ( $t_{FM} = 30.0$  nm) to  $\alpha = 0.021$  for the thinnest films ( $t_{FM} = 2.5$  nm). The inset shows the line shapes for films with and without Pt, illustrating the broadening without significant frequency shift or significant change in peak asymmetry.

A similar analysis has been carried through for CoFeB and Co (not pictured). Larger inhomogeneous linewidths are observed for pure Co, but homogeneous linewidth still exceeds inhomogeneous linewidth by a factor of three over the frequency range studied, and inhomogeneous linewidths agree within experimental error for the thinnest films with and without Pt overlayers. We extract for these films  $\alpha_0^{CoFeB} = 0.0065$  ( $G_0^{CoFeB} = 111$  Mhz) and  $\alpha_0^{Co} = 0.0085$  ( $G_0^{Co} = 234$  Mhz). The latter value is in very good agreement with the average of easy- and hard-axis values for epitaxial FCC Co films measured up to 90 Ghz,  $G_0^{Co} = 225$  Mhz.<sup>16</sup>

We isolate the effect of Pt overlayers on the damping size effect in Figure 3. Values of  $\alpha$  have been fitted for each deposited heterostructure: each FM type, at each  $t_{FM}$ , for films with and without Pt overlayers. We take the difference  $\Delta\alpha(t_{FM})$  for identical FM( $t_{FM}$ )/Cu(5nm)/Al(3nm) depositions with and without the insertion of Pt(3nm) after the Cu deposition. Data, as shown on the logarithmic plot in the main panel, are found

to obey a power law  $\Delta\alpha(t_{FM}) = Kt^n$ , with  $n = -1.04 \pm 0.06$ . This is excellent agreement with an inverse thickness dependence  $\Delta\alpha(t_{FM}) = K_{FM}/t_{FM}$ , where the prefactor clearly depends on the FM layer, highest for Py and lowest for Co. Note that efforts to extract  $\Delta\alpha(t_{FM}) = Kt^n$  without the  $FM(t_{FM})/Cu$  baselines would meet with significant errors; numerical fits to  $\alpha(t_{FM}) = Kt_{FM}^{-n}$  for the  $FM(t_{FM})/Cu/Pt$  structures yield exponents  $n \simeq 1.4$ .

Expressing now the additional Gilbert relaxation as  $\Delta G(t_{FM}) = |\gamma|M_s\Delta\alpha(t_{FM}) = |\gamma^{FM}|M_s^{FM}K_{FM}/t_{FM}$ , we plot  $\Delta G \cdot t_{FM}$  in Figure 4. We find  $\Delta G \cdot t_{Py} = 192 \pm 40$  Mhz,  $\Delta G \cdot t_{CoFeB} = 265 \pm 40$  Mhz, and  $\Delta G \cdot t_{Co} = 216 \pm 40$  Mhz. The similarity of values for  $\Delta G \cdot t_{FM}$  is in good agreement with predictions of the spin pumping model in Equation 1, given that interfacial spin mixing parameters are nearly equal in different systems.

The similarity of the  $\Delta G \cdot t_{FM}$  values for the different FM layers is, however, at odds with expectations from the "resistivity-like" mechanism. In Figure 4, *inset*, we show the dependence of  $\Delta G \cdot t_{FM}$  upon the tabulated  $\lambda_{SDL}$  of these layers from Ref<sup>17</sup>. It can be seen that  $\lambda_{SDL}^{Co}$  is roughly an order of magnitude longer than it is for the other two FM layers, Py and CoFeB, but the contribution of Pt overlayers to damping is very close to their average. Since under the resistivity mechanism, only Py and CoFeB should be susceptible to a resistivity contribution in  $\Delta\alpha(t_{FM})$ , the results imply that the contribution of Pt to the nonlocal damping size effect has a separate origin.

Finally, we compare the magnitude of the nonlocal damping size effect with that predicted by the spin pumping model in Ref.<sup>10</sup>. According to  $\Delta G \cdot t_{FM} = |\gamma|^2\hbar/4\pi = 25.69 \text{ Mhz} \cdot \text{nm}^3(g_L/2)^2(g_{eff}^{\uparrow\downarrow}/S)$ , our experimental  $\Delta G \cdot t_{FM}$  and  $g_L$  data yield effective spin mixing conductances  $g_{eff}^{\uparrow\downarrow}/S [\text{Py}/\text{Cu}/\text{Pt}] = 6.8 \text{ nm}^{-2}$ ,  $g_{eff}^{\uparrow\downarrow}/S [\text{Co}/\text{Cu}/\text{Pt}] = 7.3 \text{ nm}^{-2}$ , and  $g_{eff}^{\uparrow\downarrow}/S [\text{CoFeB}/\text{Cu}/\text{Pt}] = 9.6 \text{ nm}^{-2}$ . The Sharvin-corrected form, in the realistic limit of  $\lambda_{SDL}^N \gg t_N$ <sup>11</sup> is  $(g_{eff}^{\uparrow\downarrow}/S)^{-1} = (g_{F/N}^{\uparrow\downarrow}/S)^{-1} - \frac{1}{2}(g_{N,S}^{\uparrow\downarrow}/S)^{-1} + 2e^2\hbar^{-1}\rho t_N + (\tilde{g}_{N_1/N_2}^{\uparrow\downarrow}/S)^{-1}$ . Using conductances  $14.1\text{nm}^{-2}$  (Co/Cu),  $15.0\text{nm}^{-2}$  (Cu),  $211\text{nm}^{-2}$  (bulk  $\rho_{Cu}$ ,  $t_N = 3\text{nm}$ ),  $35 \text{ nm}^{-2}$  (Cu/Pt) would predict a theoretical  $g_{eff,th.}^{\uparrow\downarrow}/S [\text{Co}/\text{Cu}/\text{Pt}] = 14.1 \text{ nm}^{-2}$ . Reconciling theory and experiment would require an order of magnitude larger  $\rho_{Cu} \simeq 20\mu\Omega \cdot \text{cm}$ , likely not physical.

To summarize, a common methodology, controlling for damping size effects and intermixing in single films, has allowed us to compare the nonlocal damping size effect in different FM layers. We observe, for Cu/Pt overlayers, the same power law in thickness  $t^{-1.04 \pm 0.06}$ ,

the same materials independence, but roughly half the magnitude that predicted by the spin pumping theory of Tserkovnyak<sup>10</sup>. The rough independence on FM spin diffusion length, shown here for the first time, argues against a resistivity-based interpretation for the effect.

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## FIGURES

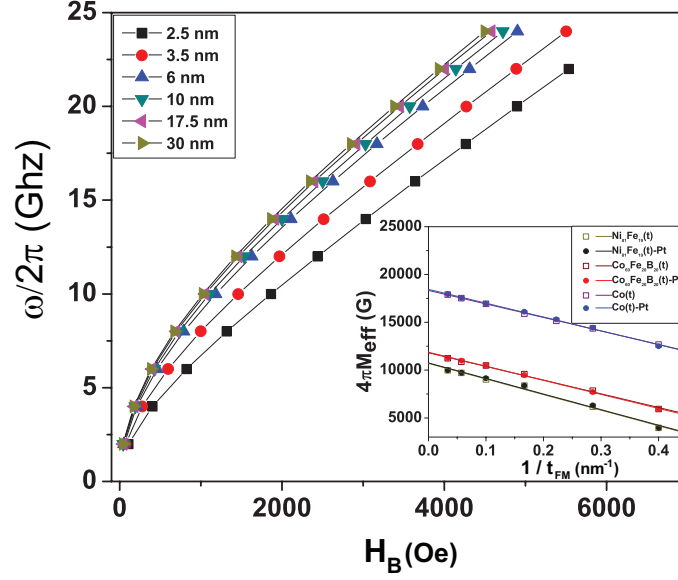


FIG. 1. Fields for resonance  $\omega(H_B)$  for in-plane FMR, FM= $Ni_{81}Fe_{19}$ ,  $2.5 \text{ nm} \leq t_{FM} \leq 30.0 \text{ nm}$ ; solid lines are Kittel fits. *Inset*:  $4\pi M_s^{eff}$  for all three FM/Cu, with and without Pt overlayers.

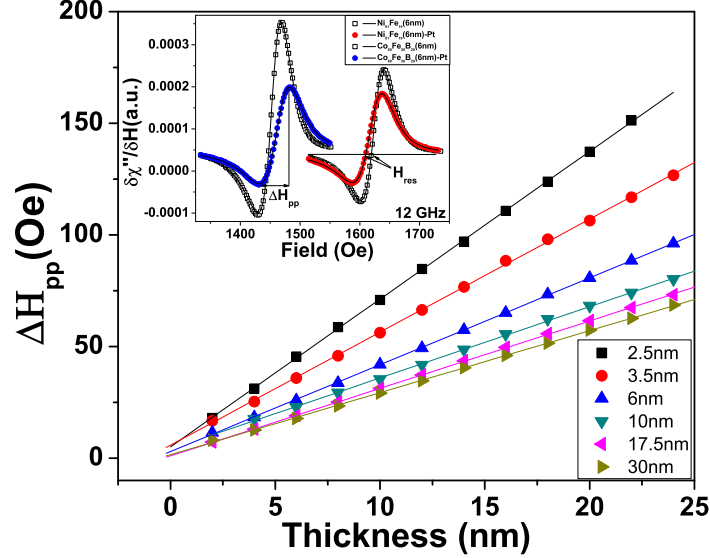


FIG. 2. Frequency-dependent peak-to-peak FMR linewidth  $\Delta H_{pp}(\omega)$  for FM= $Ni_{81}Fe_{19}$ ,  $t_{FM}$  as noted, films with Pt overlayers. *Inset*: lineshapes and fits for films with and without Pt, FM= $Ni_{81}Fe_{19}$ , CoFeB.



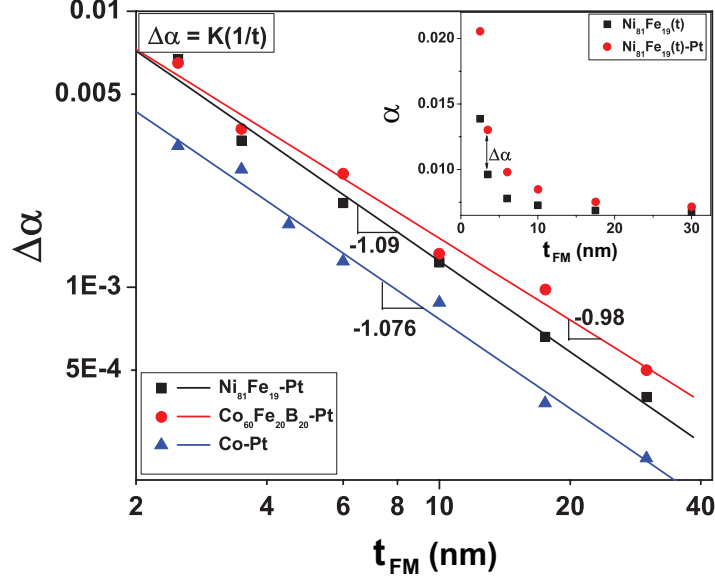


FIG. 3. *Inset:*  $\alpha_{\text{no Pt}}(t_{FM})$  and  $\alpha_{\text{Pt}}(t_{FM})$  for Py, after linear fits to data in Figure 2. Main panel:  $\Delta\alpha(t_{FM}) = \alpha_{\text{Pt}}(t_{FM}) - \alpha_{\text{no Pt}}(t_{FM})$  for Py, CoFeB, and Co. The slopes express the power law exponent  $n = -1.04 \pm 0.06$ .

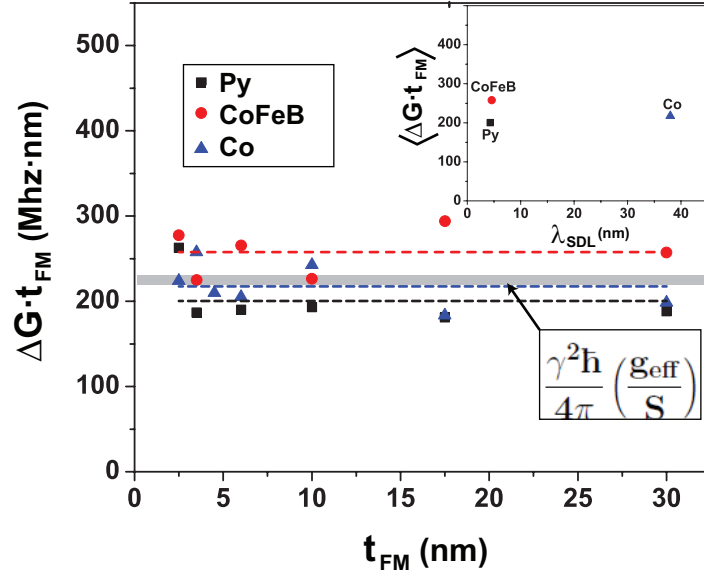


FIG. 4. The additional nonlocal relaxation due to Pt overlayers, expressed as a Gilbert relaxation rate - thickness product  $\Delta G \cdot t_{FM}$  for Py, CoFeB, and Co. *Inset:* dependence of  $\Delta G \cdot t_{FM}$  on spin diffusion length  $\lambda_{SDL}$  as tabulated in<sup>17</sup>.